Referee report:

‘Efficient use of a Lagrangian Particle Dispersion Model for atmospheric inversions using satellite observations of column mixing ratios’

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The paper describes further developments in the FLEXPART 10.4 code for simulating total column CH_4 mixing ratios. This implementation is important, as the use of satellite information is becoming increasingly valuable for identifying CH_4 hotspots and deriving emissions through inverse modelling. While the Siberia case study is interesting, and the integration of TROPOMI satellite measurements into FLEXPART is a valuable contribution, the methodology described in the manuscript is vague. It is too brief, the equations are confusing and poorly described, and there are several errors. Given that the authors claim this is the first study to calculate source-receptor relationships (SRRs) for column-averaged mixing ratios observed by satellites, a more thorough explanation of the methodology is needed. I believe the paper has potential; however, in its current form, I do not recommend it for publication unless the authors address all major comments listed below.

Major revisions:

1. As soon as I started reviewing this article, my first question was how the changes in this version of FLEXPART 10.4 differ from the recent release of FLEXPART 11. Since some of the co-authors of this paper also contributed to a recent publication - Bakels et al. (2024), which also demonstrates how FLEXPART can be used to simulate total column source-receptor relationships, it is unclear why the authors do not reference that work. For readers and new users of FLEXPART, it would be helpful if the authors explained the key differences between the two versions. Which version of the code should be used for simulating total column source-receptor relationships for satellite data?

2. In Section 2.1, the authors attempt to explain how total column observations were modelled using FLEXPART, but they do not specify which subroutines were modified. For clarity and reproducibility, it would be helpful to identify the main changes in the code. For example, mentioning specific subroutines - such as readreleases.f90, which handles the RELEASES input file - would be valuable for readers and new users. A brief description of the implemented changes, including key equations and code modifications, would enhance the utility of this work. It is possible that some of these details were presented in Bakels et al. (2024), but no reference is provided.

3. One of the main concerns is how some equations are presented in the article. For example, the authors state that the column SRR in FLEXPART 10.4 can be obtained using Equation (6).

$$H_{ijk}^{col} \sum_{n=1}^{N} a_n w_n \frac{t}{m_n \rho_{ijk}} \sum_{p=1}^{J_{i,jk,n}} m_p$$

Here, the authors describe *t* as the sampling time, but my understanding is that *t* in the equation should be interpreted as $\Delta t_{p,i,j,z \leq h}$. In other words, the sensitivity of the receptor located at x_r at time t_r to surface fluxes originating from x_i, y_j is obtained by summing the time spent by particle *p* over the grid cell *i,j* within the surface layer of height *h* at each discrete time step (see Equation 4 in Wu et al., 2018), who implemented total column simulations using a different but similar LPDM model. This is also described in Seibert and Frank (2004), Equation 8, where $\Delta t'_{i\,j\,n}$ is defined as the residence time of a trajectory in the spatio-temporal grid cell (i,n).

4. Assuming that Equation 8 is correct, shouldn't equation 9 be expressed as follow:

$$H_{ijk}^{col} = \frac{t}{m_n \, \rho_{ijk}} \sum_{n=1}^{N} \sum_{p=1}^{J_{i,j,k,n}} \frac{P_n}{P} m_p$$

It is not clear how Equation 8 can be further simplified to:

$$H_{ijk}^{col} = \frac{t}{m_n \, \rho_{ijk}} \sum_{p=1}^{J_{i,j,k,n}} m_p$$

Equation 9 requires further explanation. It is unclear how the authors transition from Equation 8 to Equation 9. They mention that Equation 9 can be seen as analogous to the calculation for point observations, but how exactly?

Later is mentioned that total column of BRR (equation 10) for the grid cell ijk can be written as:

$$H_{ijk}^{col,\,ini} = \frac{1}{m} \sum_{n=1}^{N} \sum_{p=1}^{J_{i,j,k,n}} \frac{P_n}{P} m_p$$

I would have expected Equation 10 to be similar in form to Equation 8.

$$H_{ijk}^{col,\,ini} = \frac{t}{m_n \, \rho_{ijk}} \sum_{n=1}^{N} \sum_{p=1}^{J_{i,j,k,n}} \frac{P_n}{P} m_p$$

It is not clear how the final particle position is calculated in Equation 10.

5. The paragraph from lines 130 to 135 requires further clarification. I do not fully understand the following part: "*In FLEXPART, an observation can be represented by releasing virtual particles from a volume in which the particles are distributed randomly. However, the default is that this volume is aligned with the meridians and parallels. Therefore, we have implemented an affine transformation on the particle positions so that the volume they represent matches the geometry of the retrieval*." This description is vague and confusing. It would be helpful if the authors

provided the explicit form (i.e., equations) of the affine transformation applied to the particle coordinates.

6. In section 2.2 (averaging of retrievals), the authors do not mention whether the super observations were created in a rectangular grid cell or not, or use the distorted rectangles given by the satellite grid-cells.

   Equation 12 from Section 2.2 (Averaging of retrievals) needs further review, the third term of the equation is wrong. Shouldn't be:

$$- \frac{1}{S} \sum_{m=1}^{M} \sum_{n=1}^{N} s_m a_{mn} w_{mn} x_{mn}^{pri}$$

Why the term of equation 13, cannot be further simplified as:

$$- \frac{1}{S} \sum_{n=1}^{N} \overline{a_n}\, \overline{w_m}\ \overline{x_n^{pri}}$$

7. In Section 3.1.3. it is not clear if inversion uses temporal correlation. If not, why?

8. At the beginning of the results section, and before presenting the modelled posterior XCH_4 (Section 3.2.1), it would be helpful to include a subsection showing the convergence diagnostics of the inversion, particularly given the use of TROPOMI satellite data. I recommend that the authors report the evolution of the cost function during the optimisation, along with the final cost function value normalised by the number of observations (i.e., final cost function J/n), which should ideally approach 0.5 under Gaussian error assumptions. Including these diagnostics would help assess the quality of the inversion and whether the assumed error statistics are appropriate.

9. The authors mention in Section 3.2.2 that the posterior fluxes remain very close to the prior, with a total mean posterior source of 30.3 Tg yr^{-1} compared to the prior estimate of 31.0 Tg yr^{-1}. This limited adjustment suggests that the observational constraint had minimal impact on the flux optimisation. I recommend that the authors provide an analysis of the inversion's sensitivity to prior and observation uncertainties. In particular, it would be helpful to include convergence diagnostics, as stated previously (e.g., cost function components and their evolution), to assess whether the inversion system is appropriately weighted and whether the observations are effectively contributing to the flux estimates.

10. In Section 3.2.2, it is also mentioned that the uncertainty reduction in the fluxes is quite small, indicating that the pattern of uncertainty reduction follows the distribution of observations and the prior flux uncertainty. However, if the prior uncertainties (i.e., variances in B) are too small, the inversion will not adjust the fluxes significantly, even if the observations suggest otherwise. Similarly, if the observation uncertainties (i.e., variances in R) are too large, the inversion will

down-weight the observational constraints, resulting in minimal deviation from the prior.

11. In Section 3.2.3, the authors state: "The difference between the posterior fluxes from the inversion using ground-based observations versus that using TROPOMI (Fig. 8c) follows a very similar pattern to the posterior-minus-prior flux increments (Fig. 8b), as expected, since the posterior fluxes from the inversion using TROPOMI are very close to the prior." This raises the question: are we truly learning anything from TROPOMI? As noted earlier, an analysis of the convergence diagnostics would provide greater confidence in the results. It remains unclear whether the assumed uncertainties are appropriate and whether they lead to reliable posterior flux estimates.

Minor revisions:

Please add a title to each panel in Figure 2 to clarify which maps correspond to the observations, prior, and posterior. The legend should also include units.

In Figure 1, the country names should be labelled for better reference - the text mentions China, but it is unclear which boundary the authors are referring to.

Reference:

Bakels, L., Tatsii, D., Tipka, A., Thompson, R., Dütsch, M., Blaschek, M., Seibert, P., Baier, K., Bucci, S., Cassiani, M., Eckhardt, S., Groot Zwaaftink, C., Henne, S., Kaufmann, P., Lechner, V., Maurer, C., Mulder, M. D., Pisso, I., Plach, A., Subramanian, R., Vojta, M., and Stohl, A.: FLEXPART version 11: improved accuracy, efficiency, and flexibility, Geosci. Model Dev., 17, 7595–7627, https://doi.org/10.5194/gmd-17-7595-2024, 2024.

Seibert, P. and Frank, A.: Source-receptor matrix calculation with a Lagrangian particle dispersion model in backward mode, Atmos. Chem. Phys., 4, 51–63, https://doi.org/10.5194/acp-4-51-2004, 2004.

Wu, D., Lin, J. C., Fasoli, B., Oda, T., Ye, X., Lauvaux, T., Yang, E. G., and Kort, E. A.: A Lagrangian approach towards extracting signals of urban CO2 emissions from satellite observations of atmospheric column CO2 (XCO2): X-Stochastic Time-Inverted Lagrangian Transport model ("X-STILT v1"), Geosci. Model Dev., 11, 4843–4871, https://doi.org/10.5194/gmd-11-4843-2018, 2018.